Gedik et al. Reply: In a recent Letter [1], we reported experiments in which nonequilibrium quasiparticles were generated and detected in a single crystal of YBa₂Cu₃O_{6.5} (YBCO6.5) using time-resolved optical spectroscopy. The nonequilibrium state was probed through measurements of the photoinduced change in reflectivity, ΔR , which is proportional to the nonequilibrium quasiparticle density, n. We found that the decay rate, γ , of ΔR depends on the temperature, T, and n, according to $\gamma = \gamma_{\text{th}}(T) + \beta n$. The linear dependence of γ on n demonstrates that the decay rate is controlled by the pairwise scattering of quasiparticles and that the phonon bottleneck effect, which is ubiquitous in s-wave superconductors, is absent in YBCO6.5.

The coefficient β , in principle, can provide a measure of the strength of quasiparticle interactions in cuprate superconductors. However, in order to interpret β it is first necessary to identify the quasiparticle-pair scattering process. In s-wave superconductors there is no controversy regarding this process: a pair of quasiparticles scatters into the condensate with the simultaneous emission of a phonon. Clearly, such a process can take place in a cuprate superconductor as well. However, there is an additional channel in a d-wave superconductor in which a pair can scatter with emission of lower energy (nodal) quasiparticles. In our Letter we referred to the former process as recombination, and the latter was termed thermalization. As a way of explaining the absence of a phonon bottleneck, we suggested that thermalization may be the dominant process giving rise to the decay of ΔR .

In their Comment, Demsar et~al. do not question the findings summarized above. However, they suggest that recombination, rather than thermalization, is the dominant process leading to the decay of ΔR . We agree that this is indeed a possibility provided one can understand the absence of the phonon bottleneck. However, in advocating their point of view, Demsar et~al. raise objections to aspects of data interpretation and analysis presented in our Letter. We believe that their objections are not valid and below we respond to the three points they raise.

(1) The first point concerns the sign of ΔR . We proposed that ΔR results from the shift in conductivity spectral weight from $\omega = 0$ to higher frequency as Cooper pairs are broken. We showed theoretically that this gives rise to a negative $\Delta \epsilon_1$ at the probe frequency that is proportional to the second moment of the spectral weight shift. Demsar et al. object that, for light polarized along the chain direction in YBCO, this mechanism gives the wrong sign for ΔR . However, they fail to account for the exponential decrease of $\Delta \epsilon_1$ due to absorption of the pump beam [2]. This adds an additional negative term to ΔR , modifying their formula to $\tilde{n}^2 - 3\kappa^2 - 2\kappa\kappa_p - 1$, where κ_n is the imaginary part of the index as seen by the pump. We suggest that measurements of \tilde{n} and κ in the same crystal as used to measure ΔR may be necessary to determine the sign of $\Delta \epsilon_1$.

- (2) By analyzing the dependence of ΔR on laser fluence we determined a characteristic saturation energy density, $u_{\rm sat}$, of 0.43 J/cm³. Demsar *et al.* take issue with this determination, citing their Refs. [6,7] as demonstrating that this fluence is too small to destroy superconductivity. Neither of those papers reports the change in optical response as a function of fluence, so it is difficult to judge the basis of this claim. However, independent measurements verify that 0.43 J/cm³ is indeed a reasonable value. For example, the condensation energy as determined by Loram *et al.* [3] from specific heat data is 0.3 J/cm³ which is very close to our experimental value of 0.43 J/cm³.
- (3) Demsar *et al.* argue that our proposal that the decay of ΔR reflects a thermalization process is refuted by optical pump-THz probe experiments by Averitt *et al.* (Ref. [7] of [4]). The claim is that σ_2 is a direct measure of the condensate spectral weight and that its recovery indicates rapid misinterpretation of THz spectra. In fact, σ_2 is not a direct measure of the condensate spectral weight. For $\omega > 1/\tau$ (where $1/\tau$ is the scattering rate) the quasiparticle and condensate contributions are comparable. In YBCO $1/\tau$ is less than the minimum frequency of 500 GHz reported by Averitt *et al.* Therefore this experiment cannot distinguish whether spectral weight has been restored to the condensate or to nodal quasiparticles.

The second point in (3) is that a relaxation rate proportional to excitation density is well-known in conventional *s*-wave superconductors. In fact, the situation is entirely the opposite: in *s*-wave superconductors the rate of relaxation to the ground state is independent of excitation density. Rothwarf and Taylor (Ref. [9] in [4]) explained the lack of density dependence as a consequence of the coupling between quasiparticles and phonons.

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- [1] G. P. Segre et al., Phys. Rev. Lett. 88, 137001 (2002).
- [2] I. M. Fishman et al., J. Opt. Soc. Am. B 8, 1880 (1991).
- [3] J. Loram et al., J. Phys. Chem. Solids **59**, 2091 (1998).
- [4] J. Demsar *et al.*, Phys. Rev. Lett. preceding Comment, **91**, 169701 (2003).